

ACCURATE INTEGRAL COUNTING USING MULTI-CHANNEL ANALYZERS

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Abstract

Many techniques in radionuclide metrology rely on accurate measurement of the total count rate, that is integral counting, from a detector. We present the design and results from integral counting using a live-timed multi-channel analyzer (MCA) based on a list-mode digitizer and a live-timed scalar, both with imposed, extending dead times. These systems were benchmarked against an existing NIST anti-coincidence counting system. Both systems were used to determine correction factors to an MCA that uses Gedcke-Hale dead-time corrections. Tests were performed for a large-area gas-flow proportional counter and a NaI(Tl) detector for count rates up to 80000 s^{-1} . The results were used to create correction factors suitable for alpha- and beta-particle emission rate calibrations.

Keywords: integral counting; dead time; multi-channel analyzer; MCA; radionuclide metrology

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1. Introduction

Various techniques in radionuclide metrology rely on accurate measurement of the total (integral) count rate from a detector. Examples include 2π proportional counting for calibrating α - or β -particle surface emission rates and activities (Unterweger and De Felice, 2015), 4π liquid scintillation counting (e.g. CIEMAT/NIST efficiency tracing) (Broda et al., 2007), and absolute activity measurements by γ -ray methods (e.g., $4\pi\gamma$ and sum-peak counting) (Brinkman, 1979; Thiam et al., 2015).

For metrology-level measurements (uncertainties typically below a few percent), accurate counting in these applications is often implemented using custom-built, live-timed, counting systems, which have measured accurate activities with count rates up to 10^6 s^{-1} (Bouchard, 1994; Bouchard, 2000). However, a shortcoming of these traditional systems is their scalar nature, based on a single threshold per measurement, which does not provide spectroscopic information on the energy deposited in the detector. As a result, the live-timed counting system is not adequate for identifying radionuclide impurities in a sample, setting thresholds for integral counting, or monitoring the detector response for quality. These essential tasks are often accomplished using a parallel signal chain terminating in a multichannel analyzer (MCA). For some cases, particularly for low count rates where dead time is negligible, MCAs have been used for integral counting as well, though often with separate analysis of dead-time uncertainty (Pommé et al., 2015).

This work is motivated by the need to perform 2π α/β proportional counting for calibrating surface-emission rate sources that are in turn used to calibrate or test detectors used in radiation protection with rates up to about $5 \cdot 10^4 \text{ s}^{-1}$ (corresponding to $\approx 1 \cdot 10^5 \text{ Bq}$)

by a procedure that requires extrapolation of a spectrum down to 0 keV energy (Hutchinson, 2004; King et al., 2018). We developed two methods to achieve this goal that involve using MCAs with live timing. First, we used a pulse digitizer in list-mode acquisition, with real-time data processing to implement a live-timed MCA. Second, we used a commercial MCA intended for γ -ray spectrometry that was run in parallel to a simple, custom-made live-timed counter. The two methods were validated against the well-established NIST live-timed anticoincidence counting system.

2. Equipment and Methods

The three measurement systems that were compared are shown in

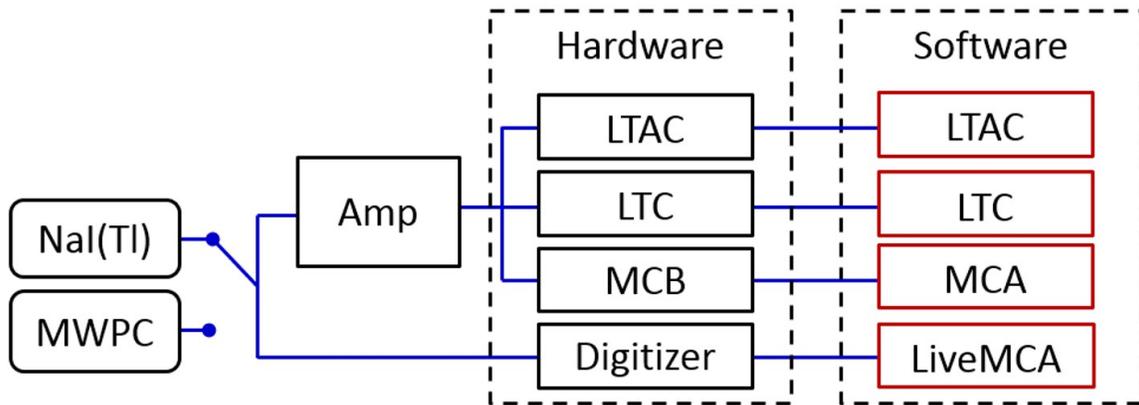


Figure 1 and referred to in this paper as the live-timed counter (LTC), multi-channel analyzer (MCA) correction and live-timed MCA (LiveMCA). The reference value for validation came from the NIST live-timed anti-coincidence system (LTAC), also shown in

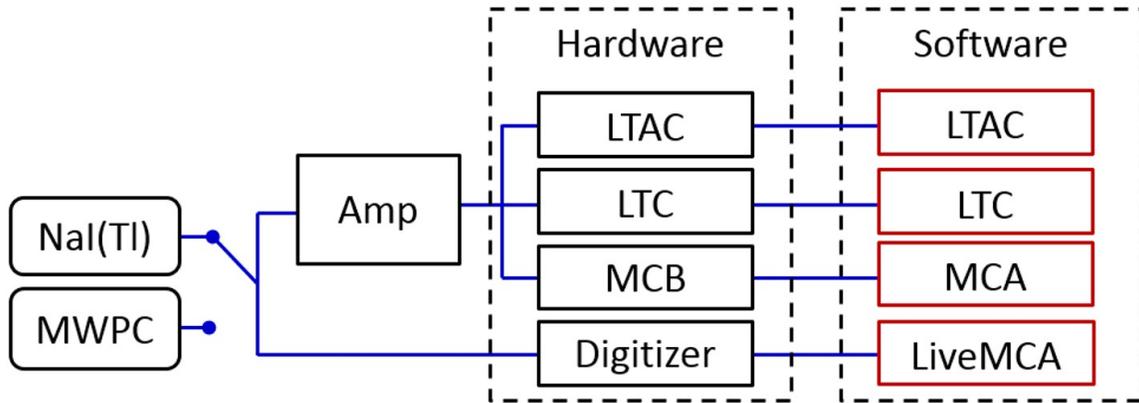


Figure 1, which has been routinely used for precision primary standardizations over the past 30 years.

Various combinations of the systems were connected in parallel to the preamplifier output signal from either a 2π gas-flow multi-wire proportional counter (MWPC) or a NaI(Tl) detector. In this way the systems could be compared while mitigating the variance due to Poisson counting statistics during a given counting period.

2.1 MWPC

The MWPC and its use for calibrating α - and β -particle emission rates and activities from large-area sources has been extensively documented (Hutchinson and Bright, 1987; Hutchinson, 2004; King et al., 2018). Such measurements require integrating the count rate in an energy spectrum above some threshold, typically followed by an extrapolation of that spectrum to zero energy.

2.2 LTAC

The NaI(Tl) detector is part of the NIST live-timed anti-coincidence counting (LTAC) system. The LTAC measurement system, which has been described in detail previously (Lucas, 1998; Fitzgerald, 2016), uses a liquid scintillation detector in anti-coincidence with a NaI(Tl) detector and employs an analog implementation of the imposed extending-deadtime (EDT) method described in **Figure 2**. Relevant to the present work, LTAC uses an ORTEC¹ 552 single channel analyzer (SCA) (Ametek, Oak Ridge, TN, USA) for counting and an ORTEC 485 Amplifier (Amp) (bipolar output, approximately 2 μ s shaping time) for output to an MCA. The LTAC system has been the workhorse for primary standardization of radionuclides at NIST and is like systems used by other National Metrology Institutes (NMIs) around the world (Bobin, 2007). The LTAC system has produced numerous primary standards with typical standard uncertainties of 0.3 % to 1.0 %, and these primary standards have been shown to agree with those from other NMIs through international Key Comparisons in the International Reference System (SIR) (Michotte and Fitzgerald, 2010), as documented more generally in the Key Comparison Database (KCDB) maintained by the International Bureau of Weights and Measures (BIPM) (BIPM, 2019).

2.3 LTC

The LTC was designed to be a simple, low-cost, and easily implemented live-timed counter for a single channel. The LTC hardware consists of an off-the-shelf single channel analyzer (SCA) (ORTEC 550 SCA; Ametek, Oak Ridge, TN, USA) and data

¹ Certain commercial equipment, instruments, or materials are identified in this paper to foster understanding. Such identification does not imply recommendation by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

acquisition (DAQ) module (NI-6341, National Instruments, Austin TX, USA). An amplified detector signal is input to the SCA, which also receives a threshold setting from the DAQ. The SCA logic output is connected to one of the DAQ digital input lines, which is set to record time intervals between successive events. The DAQ has a 100 MHz oscillator and 32-bit counter, which is used to measure time intervals of duration up to 43 s without rolling over. Communication to the DAQ module was carried out by USB using either C code or LabView (National Instruments, Austin TX, USA). The final LTC software, developed in LabView, allows the user to set the SCA threshold, such that it is equivalent to that on a commercial MCA operated in parallel, for comparing count rates for comparable regions of interest.

The LTC live-time analysis is performed in software by implementing the imposed extending-dead-time method used by LTAC. The imposed dead time is chosen to be longer than the pulse width or any intrinsic deadtime of the system (Baerg, 1976). The extending nature of the applied dead time accounts for pulses that occur during a previous dead-time interval. The basic algorithm is expressed in **Figure 2**.

2.4 MCA

The MCA system consisted of an ORTEC ASPEC-927 Multichannel Buffer (MCB) with accompanying MCA emulation software, one of various commercial products that similarly use Gedcke-Hale dead-time correction (Jenkins et al., 1981), which corrects for dead time over a period defined by the amplifier output, rather than for an external, imposed period. This method is used for x-ray and γ -ray spectroscopy (Pommé and Kennedy, 2000). The MCA inputs are the energy signal and optional busy signal from the amplifier. The user threshold for the MCA was set and then scaled to set an equivalent threshold in the LTC.

The LiveMCA hardware was a CAEN DT 5724 (CAEN S.p.A, Viareggio LU, Italy) digitizer, which outputs list-mode data for each event as a pulse height and time stamp. The digitizer input was the preamplifier output of either the MWPC or the NaI(Tl) detector. The pulse-processing software was developed in-house using LabView to create a live-timed MCA based on the algorithm in **Figure 2**, with the additional component of binning the counts by energy. In this way, the advantages of live-timing with imposed extending-dead time could be combined with those of an energy spectrum.

2.5 Sources

The radioactive sources used with the MWPC were 6 commercial large-area, anodized, alpha-particle emission sources, 5 of Pu-238 and 1 of Am-241 (Eckert and Ziegler, Berlin Germany). No absolute calibration was used for the emission rate of the sources. Instead the sources were used to produce consistent output count rates from the MWPC, which then fed in to multiple data acquisition systems in parallel. The commercial α -emission

sources were not used as calibrated standards to avoid circular logic that would be caused by the fact that the MWPC is the means of source calibrations.

The absolute reference came from NaI(Tl) measurements on a set of 4 Co-57 point sources that were prepared previously by gravimetric dispensing of 3 gravimetrically-linked dilutions of a master solution. At the measurement time, the activities of the 4 sources ranged from $2.2 \cdot 10^4$ Bq to $1.1 \cdot 10^6$ Bq. The relative γ -ray emission rates of the various sources were known to better than 0.1 % from standard gravimetric dispensing (Sibbens and Altzitzoglou, 2007), therefore the linearity of the counting systems could be compared over a wide range of count rates. The reference value for the true count rate was taken from the LTAC system at low count rates ($\approx 1 \cdot 10^3$ s⁻¹), typical of primary standard measurements. To fill in a wide range of known count rates in the NaI(Tl), the sources were measured at 2 source-to-detector distances of 28 mm and 75 mm.

A commercial Cs-137 γ -ray point source (Eckert and Ziegler, Berlin Germany) of activity $\approx 2 \cdot 10^5$ Bq was used for further head-to-head comparisons of multiple counting systems.

3. Results and Discussion

Measurements are reported with the following nomenclature. The observed count rate is R_{obs} , the estimated value for the true count rate is R . A correction factor from R_{obs} to R is defined as,

$$f = \frac{R}{R_{obs}}. \quad (1)$$

The counting accuracy of the LTAC system has been verified by multiple methods, including pulser tests and the decaying-source method. Here we report a simultaneous test of LTAC, LiveMCA and MCA by measuring a set of gravimetrically-linked Co-57 point sources. The true value was taken as the measured value using one channel of the LTAC counter for count rates of about $1.6 \cdot 10^3 \text{ s}^{-1}$, with EDT of $20 \mu\text{s}$, which are typical values for LTAC measurements. Count times were 300 s to 600 s, to achieve $> 5 \cdot 10^5$ counts. Corrections for background and radioactive decay were applied prior to comparing results, shown in

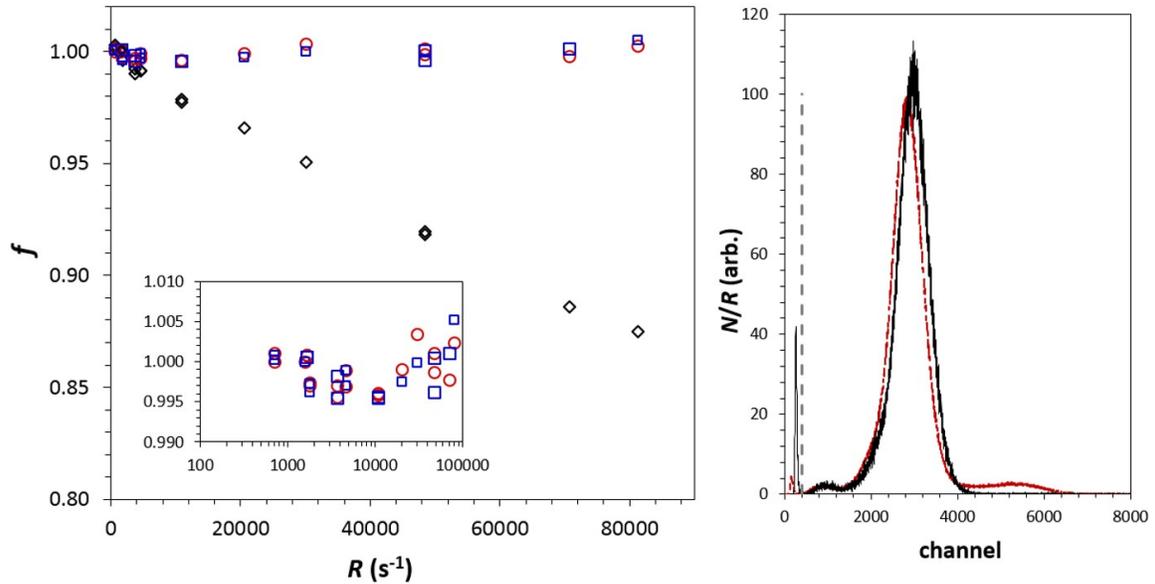


Figure 3. The mean difference and standard deviation between LiveMCA and LTAC, run in parallel, for $R_{obs} \leq 8.0 \cdot 10^4 \text{ s}^{-1}$ was $(0.03 \pm 0.14) \%$. Neither system showed a significant correlation of f vs. R_{obs} . The standard deviation of f for repeats of similar R_{obs} values was 0.18% for both systems, consistent with counting statistics. A small curvature can be seen in the data, which may be caused by spectral changes near the low-

energy threshold. In contrast, MCA deviated from the other two significantly, with $f=0.875$ at $R_{\text{obs}}=8.0 \cdot 10^4 \text{ s}^{-1}$, with standard deviation of 0.33 % for repeats of similar R_{obs} values.

In a separate measurement, LTC was compared against LTAC with a single Cs-137 point source located at various distances and producing count rates up to $3.2 \cdot 10^4 \text{ s}^{-1}$. In addition, LTC was run for a series of EDT settings and a minimum value of 5 μs was found to be necessary, with a bias of -1.5 % at 2 μs EDT for a true rate of $3.2 \cdot 10^4 \text{ s}^{-1}$. The difference in live count rate for the two systems at 20 μs EDT was < 0.02 % for all rates tested, as expected since both systems use the same methodology and were previously validated with Co-57. The Cs-137 and Co-57 results combine to demonstrate the equivalence of LTAC, LTC, and LiveMCA for integral counting.

For the MWPC measurements, various combinations of LTC and MCA were run in parallel with matched thresholds. The MWPC is typically run with a long amplifier shaping time of 3 μs , due to the long detector pulse time produced by this large detector. To test whether the discrepancy of the MCA from the other systems could be reduced, shorter shaping times were also tested. For $R = 2.4 \cdot 10^4 \text{ s}^{-1}$, shaping times of 0.5 μs , 1.0 μs , and 3.0 μs resulted in f values of 0.969, 0.962, and 0.927, respectively, indicating a tradeoff between full integration of the detector pulse and accurate integral counting by this system.

To enable operation of the MCA at 3 μs shaping time, f values were measured for MCA using LTC with matched thresholds,

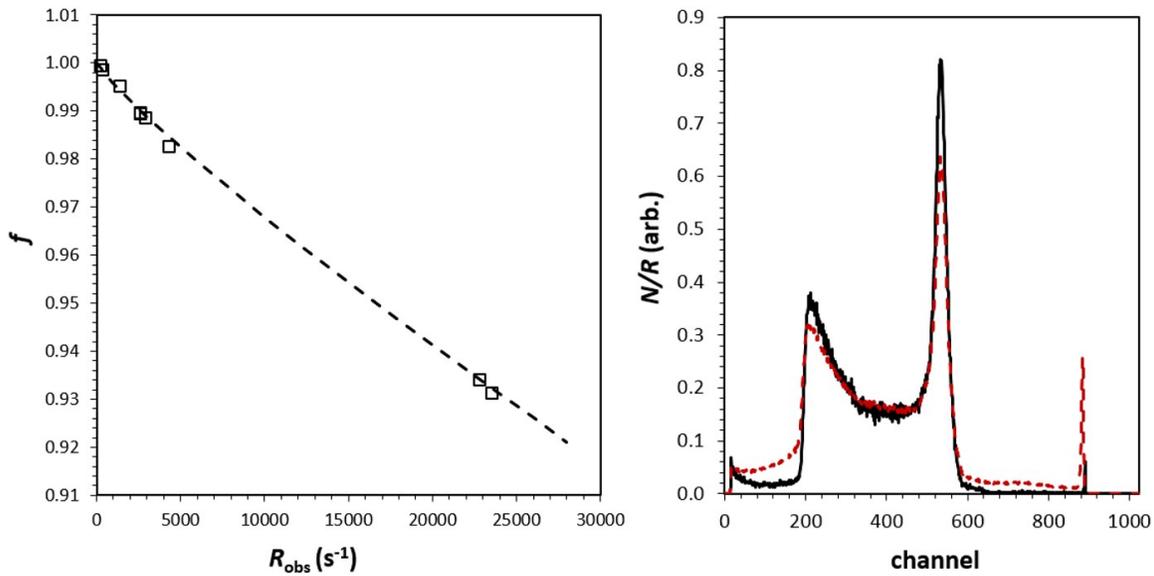


Figure 4.

4. Conclusion

We demonstrated the accuracy of integral counting for rates up to $8 \cdot 10^5 \text{ s}^{-1}$ using a simple live-timed counter (LTC) with an adjustable, imposed extending dead time, validated using a single channel of a more-complicated NIST live-timed anti-coincidence counting system and a set of gravimetrically-linked γ -ray sources. The LTC is designed to be operated in parallel with a commercial MCA such that the MCA can be used for spectral inspection and extrapolation of the integral count rate to zero energy, with the LTC providing absolute integral count rate. This combined measurement can be done during source calibrations, or periodically to calculate correction factors to the MCA count rates. Also, we demonstrated a live-timed MCA which leverages a modern pulse digitizer and software-applied imposed, adjustable, extending-dead time to achieve high-accuracy integral counting with spectral analysis capabilities.

The significant corrections needed for an MCA using Gedcke-Hale dead time may only be necessary for integral counting, rather than for the more typical application of γ -ray spectrometry. For the latter, pileup pulses that are not rejected may end up outside of the peak of interest and therefore may not be counted. In contrast, integral counting includes any pileup pulses that remain (displaced) in the spectrum and therefore may over-count. Such overcounting would be consistent with the correction factors for the MCA which were all found to be less than 1. Not surprisingly, the correction factors depend on the shaping time of the amplifier that feeds the MCA. Therefore, if correction factors are to be measured separately from routine MCA-based calibrations, care should be taken to ensure that the specific MCA, electronics, and electronics settings that could affect the pulse shape remain constant.

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FIGURES

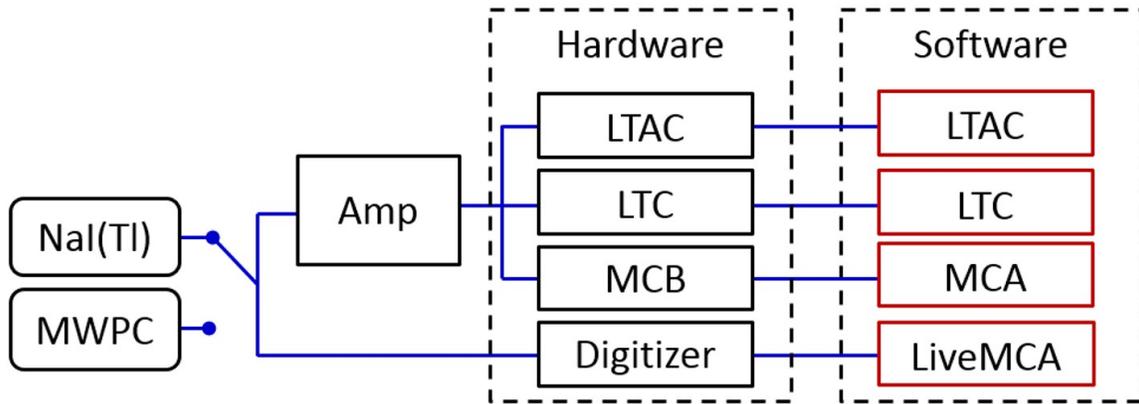


Figure 1 Concept of experimental arrangements. Abbreviations are defined in the text. Output signals from detector preamplifiers are shown. Not all data acquisition systems were run in parallel at the same time. The four systems are referred to by the software names throughout the paper.

```
++Real_Counts;  
If (dt > EDT)  
{  
    Live_Time += dt - EDT;  
    ++Live_Counts;  
}
```

Figure 2 Pseudo-code for live-time counting with imposed extending dead time (EDT), used by LTAC, LTC and LiveMCA. For each time interval, dt , between registered events the number of real counts is incremented. Only if dt exceeds the set extending dead time (EDT) is the number of live counts incremented and the live time increased.

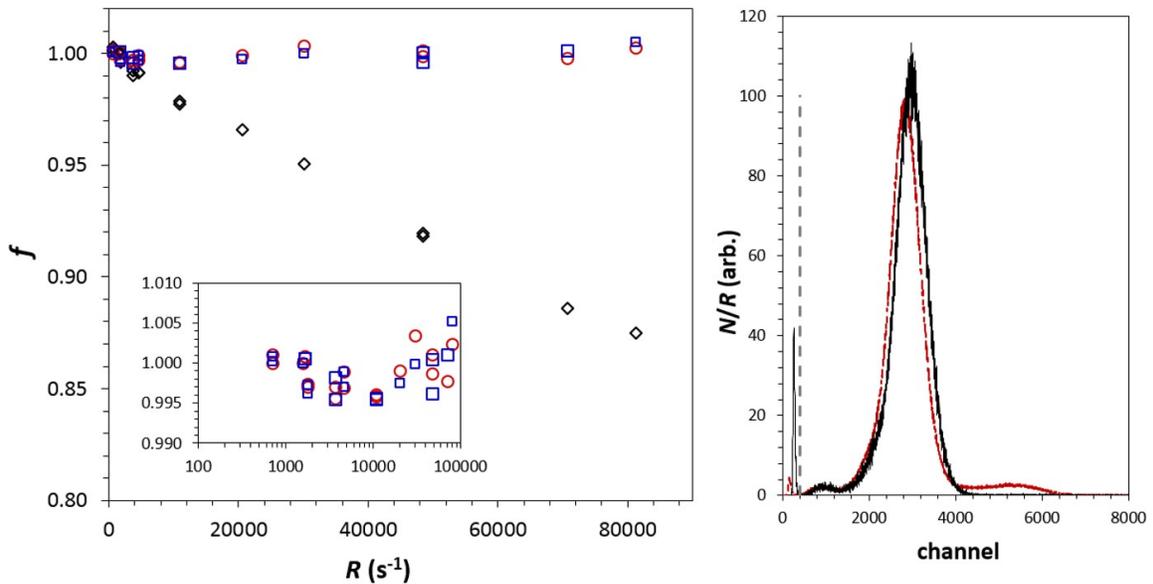


Figure 3 Left: Results for tests of LTAC, LiveMCA, and MCA for a NaI(Tl) detector using a set of gravimetrically-linked Co-57 point sources. Left plot shows correction factor, f , vs. observed count rate, R_{obs} , for LTAC (blue squares), LiveMCA (red circles) and MCA (black diamonds). Repeated points illustrate typical repeatability, consistent with Poisson counting statistics. Inset omits MCA. Right: Normalized LiveMCA spectra for $1.6 \cdot 10^3 s^{-1}$ (solid black line) and $4.9 \cdot 10^4 s^{-1}$ (dashed red line). Threshold for counting is shown by dashed gray line. Pileup and gain shift are visible for the higher R data.

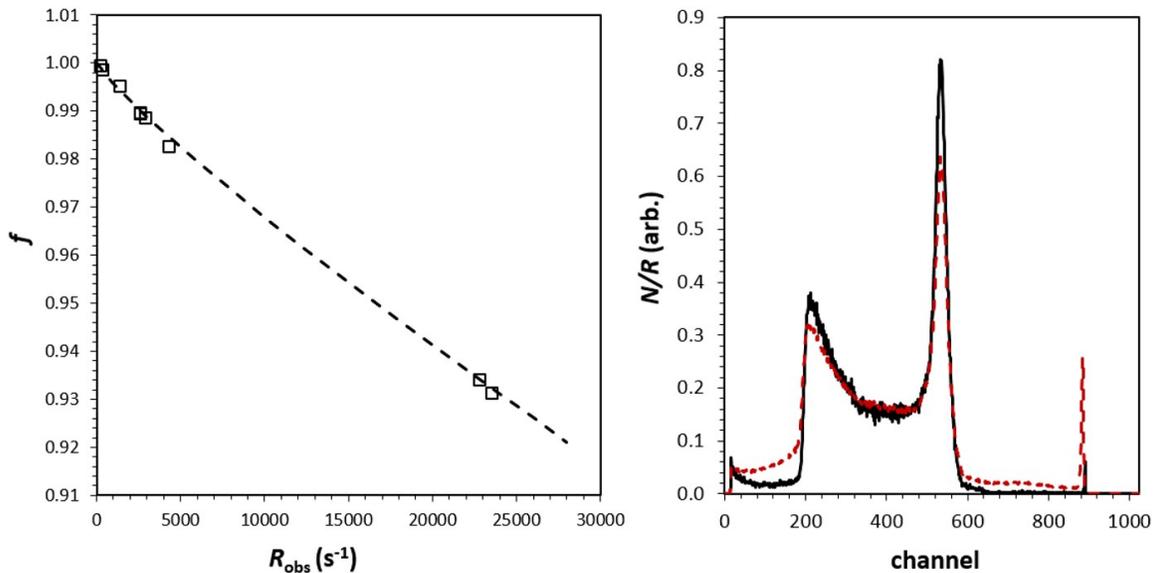


Figure 4 Left: Correction factor for MWPC with MCA for $3 \mu s$ shaping time. Right: MCA spectra showing counts per channel (N) for 100 s of live time, normalized by the corrected total count rate, R , for a low-rate source (solid black line) ($2.6 \cdot 10^3 s^{-1}$) and high rate source (dashed red line) ($2.1 \cdot 10^4 s^{-1}$). The pileup of counts into the amplifier saturation peak on right and change in shape of low-energy spectrum on left are visible. Note, the secondary peak located around channel 200 is due α -particles that do not stop in the active area of the detector (King et al., 2018).